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Responses to Reviewer #1

The authors used the CESM model with a source-tagging method to quantify the source attributions for BC direct radiative forcing (DRF) and concentration as well as polluted events. They found that in addition to regional emissions within China, emissions outside China also contribute to a large portion of BC DRF over China. This study could improve the understanding on BC pollution in China and provide implications for policy makers. Before this manuscript can be considered for publication, I have a few comments that need to be addressed by the authors.

1. One critical factor influencing BC direct radiative effect is its optical properties (absorption and extinction cross section, asymmetry factor, and single scattering albedo). Recent studies (e.g., He et al. 2015, 2016b) showed that BC optical properties vary significantly (by up to more than a factor of two) due to different coating structures and aging stages during BC aging process, which further affects direct radiative effect. (1) Could the authors add some discussions on this aspect with reference to these recent studies, for example, potential uncertainty in their results caused by this factor? (2) Could this variation in BC optical properties due to coating structures contribute to the model biases in BC AAOD simulations as discussed in the first paragraph of Section 3.2? (3) It would be helpful if the authors could add more details on how the MAM3 model computes BC optical properties. For example, does it assume a core-shell structure for internally mixed BC?
References:
He, C., et al. : Variation of the radiative properties during black carbon aging: theoretical and experimental intercomparison, Atmos. Chem. Phys., 15, 11967-11980, doi:10.5194/acp-15-11967-2015, 2015.
He, C., et al. : Intercomparison of the GOS approach, superposition T-matrix method, and laboratory measurements for black carbon optical properties during aging, J. Quant. Spectrosc. Radiat. Transf., 184, 287–296, doi:10.1016/j.jqsrt.2016.08.004, 2016b.

Response:

(1) Thanks for the suggestion. We have added discussions of the influence of aging processes on BC optical properties to the Conclusions and Discussions section, along with the references provided by the referee, as “BC aging in the atmosphere is important for BC concentration and its optical properties, which transforms BC from hydrophobic aggregates to hydrophilic particles coated with soluble materials. He et al. (2015, 2016a) found that BC optical properties varied by a factor of two or more due to different coating structures during BC aging process based on their theoretical and

experimental intercomparison. Oshima et al. (2009) and He et al. (2016b) pointed out that the use of various microphysical BC aging schemes could significantly improve simulations of BC concentrations compared to the simplified aging parameterizations. Liu et al. (2012) also reported that the wet removal rate of BC simulated in standard CAM5 is 60% higher than AeroCom multi-model mean due to the rapid or instantaneous aging of BC. H. Wang et al. (2013) showed that the explicit treatment of BC aging process with slow aging assumptions in CAM5 could significantly increase BC lifetime and the efficiency of BC long-range transport. In the three-mode aerosol module (MAM3) of CAM5 used in this study, the aging process of BC is neglected by assuming the immediate internal mixing of BC with other aerosol species in the same mode. This assumption could lead to an overestimation of wet removal of BC and, therefore, an underestimation of BC concentrations, absorption optical depth (Fig. 3) and direct radiative forcing. In addition, the internally-mixed optical treatment in CAM5 could also cause bias in BC absorption calculation. However, H. Wang et al. (2014) examined source-receptor relationships for BC under the different BC aging assumptions and found that the quantitative source attributions varied slightly while the qualitative source-receptor relationships still hold. Therefore, although the magnitude of simulated BC and its optical properties could be underestimated due to the instantaneous aging of BC and uncertainty in coating structures, we expect that the aging treatment in MAM3 of CAM5 should not influence the qualitative source attributions examined in this study."

(2) We agree that uncertainties in BC optical properties due to coating structures and/or aging could contribute to model biases in simulated BC concentrations and AAOD. We have added this statement in the discussion section. Please see the revisions above.

(3) In the MAM3 aerosol module of CAM5 used in this study, the aging process of BC is neglected by assuming an instantaneous mixing of BC with other aerosol species in the same accumulation mode, which has been added in the discussion section. We also add a more detailed description of the calculation of aerosol optical properties in the Methods section, as "Aerosol optical properties for each mode are parameterized according to Ghan and Zaveri (2007). Refractive indices for aerosols are taken from the OPAC (optical properties for aerosols and clouds) software package (Koepke and Schult, 1998), but for BC at solar wavelengths the values are updated from Bond and Bergstrom (2006)."

2. For BC emissions, a number of global and regional emission inventories have been developed, which showed large uncertainties and differences among each other (e.g., Fig. 4 in Wang et al., 2014). It would be helpful if the authors could discuss the uncertainty associated with the emission inventory used in this study and how this inventory compares with previous ones for both inside and outside China, since the authors pointed out that emissions outside

China also contribute a lot to BC DRF in China.

Reference:

Wang, R., et al. : Trend in Global Black Carbon Emissions from 1960 to 2007, Environ. Sci. Technol., 48, 6780−6787, doi: 10.1021/es5021422, 2014.

Response:

Uncertainty in China BC emissions has been estimated as –43% to 93% by Lu et al. (2011), –50% to 164% by Qin and Xie (2012), ±176% by Kurokawa et al. (2013), and –28 to 126% by Zhao et al. (2013). The BC emissions estimates used here for China in 2010 are 40% higher than those of Zhao et al. (2013) and Lu et al. (2011) and 30% higher than Klimont et al. (2016), in large part due to a higher estimate of BC emissions from coal coke production. Emissions from coke production are particularly uncertain given that "there are no measurements for $PM_{2.5}$ and BC emissions" (Huo et al. 2012) available to guide inventory estimates. Total rest of the world emissions other than China, which appear to be a major contributor to burdens over western regions, are within 1% of those from Klimont et al. (2016). We have added these discussions in Conclusions and discussions section.

We have added Table S1 to compare the anthropogenic emissions used in this study with emissions from some previous studies. The anthropogenic emissions of BC in China in 2010–2014 are larger than those used in the previous studies for earlier years, partly as a result of a higher estimate of BC emissions from coal coking production. The higher emissions likely lead to higher concentrations and direct radiative forcing, and source contributions of BC in China, compared to the values reported in these studies. We have added these descriptions in the Methods section.

We also revised Fig. 1 to include BC emissions from outside China. Emissions at regional scale are summarized here instead of Country level because the model revolution is a bit course to characterize emissions by countries. Total BC emissions from neighboring regions including rest of East Asia (REA, with China excluded), South Asia (SAS), Southeast Asia (SEA), and Russia/Belarussia/Ukraine (RBU) are shown in Figure 1c. These source regions outside China are consistent with source regions defined in the second phase of Hemispheric Transport of Air Pollution (HTAP2). South Asia and Southeast Asia have relatively high emissions. They may dominate the contribution to concentrations and direct radiative forcing of BC in China, especially southern and western China, from foreign sources through long-range transport. We have also added these in the Methods section. We did not compare the emissions outside China with other studies, which is beyond the scope of this study. However, the comparison of CEDS emissions with other emission inventories can be found in Hoesly et al. (2017), which includes detailed information of the CEDS emissions and will be submit very soon.

Table S1. Comparison of CEDS annual mean anthropogenic BC emissions in China with those used in other studies

	Year	Anthropogenic emission in China (Gg/yr)
CEDS (Hoesly et al., 2016; this study)	2010–2014	2467
MIX (Li et al., 2017)	2010	1765
HTAP V2.2 (Janssens- Maenhout et al., 2015)	2010	1741
Lu et al. (2011)	2010	1751
Qin and Xie (2012)	2009	1764
Wang et al. (2012)	2007	1879
INTEX-B (Zhang et al., 2009)	2006	1811

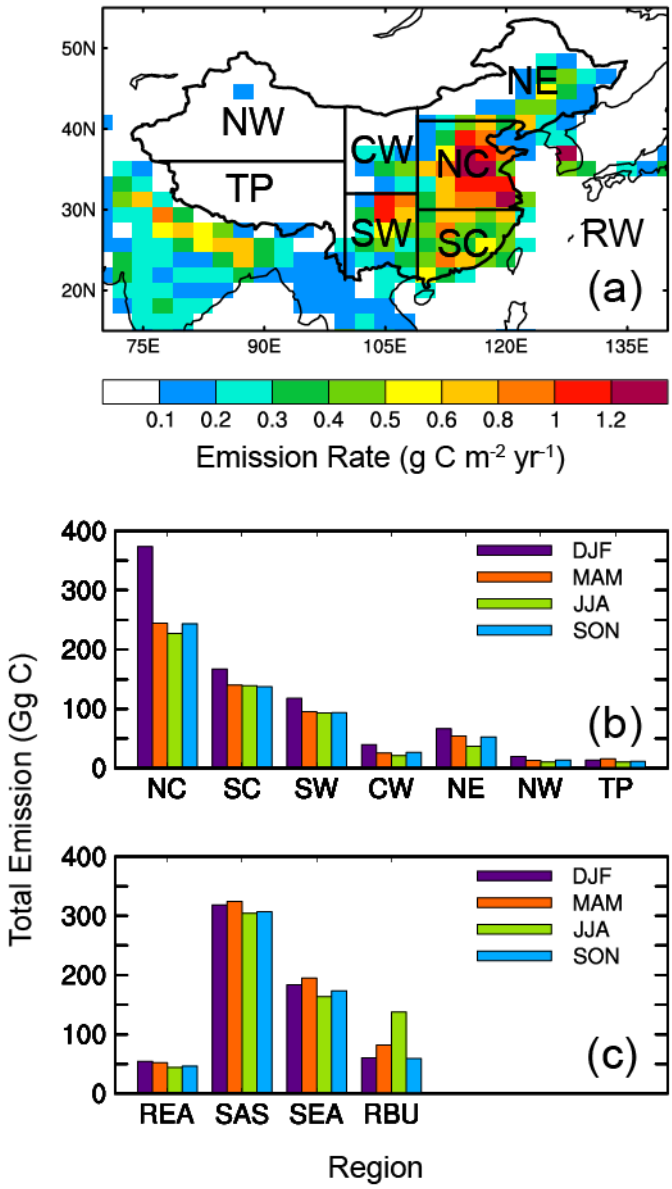

Figure 1. (a) Spatial distribution of annual mean total emissions (anthropogenic plus biomass burning, units: g C m^{-2} yr^{-1}) of black carbon (BC) averaged over 2010–2014. The geographical BC source regions are selected as North China (NC, 109°E–east boundary, 30°–41°N), South China (SC, 109°E–east boundary, south boundary–30°N), Southwest China (SW, 100°–109°N, south boundary–32°N), Central-West China (CW, 100°–109°N, 32°N–north boundary), Northeast China (NE, 109°E–east boundary, 41°N–north boundary), Northwest China (NW, west boundary–100°E, 36°N–north boundary), and Tibetan Plateau (TP, west boundary–100°E, south boundary–36°N) in China and regions outside of China (RW, rest of the world). (b) Seasonal mean total emissions (units: Gg C, Gg = 10^9g) of BC from the seven BC source regions in China and (c) emissions from rest of East Asia (REA, with China excluded), South Asia (SAS), Southeast Asia (SEA), and Russia/Belarussia/Ukraine (RBU).

3. Another important factor affecting BC simulations is aging process, which directly alters BC wet scavenging and lifetime. As pointed out by some recent studies (e.g., Oshima et al., 2009; He et al., 2016a), applying microphysical BC aging schemes could significantly improve simulations of BC concentrations compared with simplified aging parameterizations. (1) Could the authors briefly describe how BC aging is treated/computed in their model? (2) It would be helpful if the authors could briefly discuss the BC aging effect on their results with reference to these recent studies. (3) The authors mentioned in Lines 255–256 that model biases in BC concentration over China is likely due to inaccurate emissions and wet scavenging. Could this bias also be caused by model uncertainty related to BC aging? Some discussions would be useful.

reference:

He, C., Li, Q., Liou, K.-N., Qi, L., Tao, S., and Schwarz, J. P.: Microphysics-based black carbon aging in a global CTM: constraints from HIPPO observations and implications for global black carbon budget, Atmos. Chem. Phys., 16, 3077-3098, doi:10.5194/acp-16-3077-2016, 2016a.

Oshima, N., et al. : Aging of black carbon in outflow from anthropogenic sources using a mixing state resolved model: Model development and evaluation, J. Geophys. Res., 114, D06210, doi:10.1029/2008JD010680, 2009.

Response:

(1) Thanks for the suggestion. We had now added more description of the BC aging treatment and related discussions to the paper. Please see the response to comment #1 above.

(2) Following the referee's suggestion, we had now added more discussions in this regard. Please see the response to comment #1 above.

(3) Yes, the overestimation of wet scavenging is partly caused by the assumption of instantaneous aging of BC in the model. We have added the message to the manuscript, as "Larger wet removal rate and shorter lifetime of aerosols along with the instantaneous aging of BC in the MAM3 can also lead to the lower concentrations of BC (e.g., Wang et al., 2011; Liu et al., 2012; H. Wang et al., 2013; Kristiansen et al., 2016)." and in discussion section as "This assumption could lead to an overestimation of wet removal of BC and, therefore, an underestimation of BC concentrations, absorption optical depth (Fig. 3) and direct radiative forcing."

4. The authors derived BC AAOD from AERONET observations by using the method in Bond et al. (2013). However, a recent study by Schuster et al. (2016)

pointed out some weaknesses and problems related to the Bond et al. (2013) method. Could the author briefly discuss this issue? How would this affect the results in this study?

Reference:
Schuster, G. L., et al. : Remote sensing of soot carbon – Part 2: Understanding the absorption Ångström exponent, Atmos. Chem. Phys., 16, 1587–1602, doi:10.5194/acp-16-1587-2016, 2016.

Response:

We have included a discussion of this caveat associated with the BC AAOD comparison, as "Note that, the observed AAOD of BC is derived from AERONET measurements using the absorption Ångström exponent. A recent study (Schuster et al., 2016) reported that absorption Ångström exponent is not a robust parameter for separating out carbonaceous absorption in the AERONET database, which could cause biases in the AAOD estimates."

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Ghan, S. J., and R. A. Zaveri (2007), Parameterization of optical properties for hydrated internally mixed aerosol, J. Geophys. Res., 112, D10201, doi:10.1029/2006JD007927.

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Koepke, M. H. P., and I. Schult, Optical properties of aerosols and clouds: The software package opac, Bull. Am. Meteorol. Soc., 79, 831–844, 1998, doi:10.1175/1520-0477(1998)079<0831:OPOAAC>2.0.CO;2.

Wang, M., S. Ghan, M. Ovchinnikov, X. Liu, R. Easter, E. Kassianov, Y. Qian, and H. Morrison (2011), Aerosol indirect effects in a multi-scale aerosol-climate model PNNL-MMF, Atmos. Chem. Phys., 11, 5431–5455, doi:10.5194/acp-11-5431-2011.

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Qin, Y. and Xie, S. D.: Spatial and temporal variation of anthropogenic black carbon emissions in China for the period 1980–2009, Atmos Chem Phys, 12(11), 4825–4841, doi:10.5194/acp-12-4825-2012, 2012.

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